

Exponential Projectile Charge Dependence of Ar *K* and Ne *K* X-Ray Production by Fast, Highly Ionized Argon Beams in Thin Neon Targets*

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Yields of projectile and target *K* x rays from collisions between ~ 80 -MeV argon ions and thin neon targets increase dramatically as the projectile charge is increased from +6 to +17. Although incrementing the projectile charge state amounts to "tuning" projectile *L*-shell energy through target *K*-shell energy, the 60-fold (\sim exponential) increase in target yield and the more than 1000-fold ($>$ exponential) increase in projectile yield can be partially understood as the effect of a rapidly changing fluorescence yield, but not as an energy-level-matching effect.

In a recent Letter¹ we reported the observation of neon *K* x-ray yields which were strongly dependent upon the charge state of the incident highly stripped argon ion. The target x-ray yield increased by a factor of ≥ 10 when a thin foil was placed just upstream from the target, thereby changing the projectile charge from +6 to an average value of +14. Similar, but smaller, projectile structure effects have been reported earlier for light ions on heavy gas targets² and most recently for oxygen ions on solid aluminum targets.³ The new developments described here are the discovery of significantly larger projectile charge-state-dependent *K* x-ray yields than previously encountered,^{1,2} the observation of an even larger effect on the projectile yield than on the target yield, and a systematic test of the energy-level-matching hypothesis at relatively high projectile velocity.

The binding energy of an electron that is excited into the normally empty *2p* shell of Ar¹⁴⁺ is nearly equal to that of a *1s* electron in the neutral neon atom. This near match of energy levels suggested the possibility of an enhanced cross section for electron transfer from the neon *K* shell to the empty Ar¹⁴⁺ *L* shell during deeply penetrating collisions. Such an electron promotion presumably cannot occur when the projectile is Ar⁶⁺ because in that case the *L* shell is full.

In order to investigate this level-matching hypothesis, we have improved the experiment by interposing an analyzing magnet between stripping foil and gas cell which is capable of select-

ing, at 80 MeV, usable argon currents in single charge states +12 through +17 covering the range from carbonlike to hydrogenlike argon. The primary beam Ar⁶⁺ is also available when the foil is removed. The yield of *both* target and projectile *K* x rays is found to increase dramatically with projectile charge state throughout the region studied. An overall 60-fold (exponential) increase in neon *K* x-ray yield is observed together with more than a 1000-fold ($>$ exponential) increase in the argon *K* x-ray yield when the argon charge is varied from +6 to +17. As far as is known, these are the largest projectile charge-state-dependent effects ever reported for *K* x-ray production. No particular enhancement occurs in the region where energy-level matching is expected to occur, which implies that level matching is not an important criterion for this ionization process at this velocity. The new results demonstrate even more emphatically than before the need to incorporate detailed projectile structure into theories of inner-shell ionization.

With the exception of the intervening charge-state-selecting magnet mentioned above, the apparatus is little changed from that described in Ref. 1. The beam energy of 78.2 MeV corresponds to a projectile velocity 1.94×10^9 cm/sec that is slightly less than $Z_{\text{target}}\alpha c = 2.19 \times 10^9$ cm/sec, a velocity approximately equal to that of a neon *K*-shell electron. Gas-cell pressures were kept low enough (~ 20 mTorr) that few projectiles made more than one collision in the target. This fact was verified by a measured linear de-

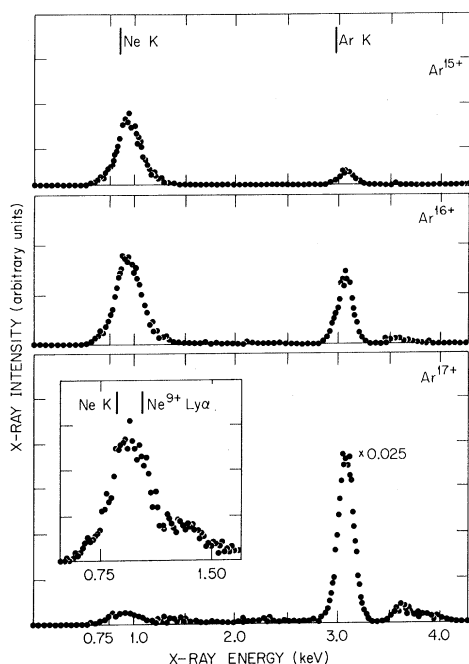


FIG. 1. X-ray spectra resulting from collisions of 80-MeV Ar ions with Ne. The electronic resolution is 150 eV. The Si(Li) detector window and dead layers attenuate the neon peak by a factor of ~ 10 compared with the argon peak.

pendence of x-ray yield on target gas pressure. A conservative estimate of electron capture cross section ($\sigma_{\text{capt}} \approx 10^{-16} \text{ cm}^2$) implies $\approx 6\%$ of the beam undergoes charge exchange in the target. The experimental results are summarized in the figures. Figure 1 gives a qualitative impression of the increase in projectile and target K x-ray yields with increasing projectile charge state. These spectra have been normalized to the same gas cell pressure and the same number of incident particles. They have not been corrected for detector-window absorption, which is approximately 10 times greater for the neon line than for the argon line. Note that at low projectile charge states the neon K x-ray production dominates the collision process, whereas at the highest projectile charge state studied the argon K x-ray production dominates. The neon peak is much broader than the argon peak in all cases, which suggests that the neon peak contains unresolved lines from a significantly greater spread of charge states than does the argon peak. The most highly ionized projectiles greatly excite the target (see inset) causing a centroid shift of some 60 eV and the appearance of (unresolved) lines whose energies correspond to those expected for

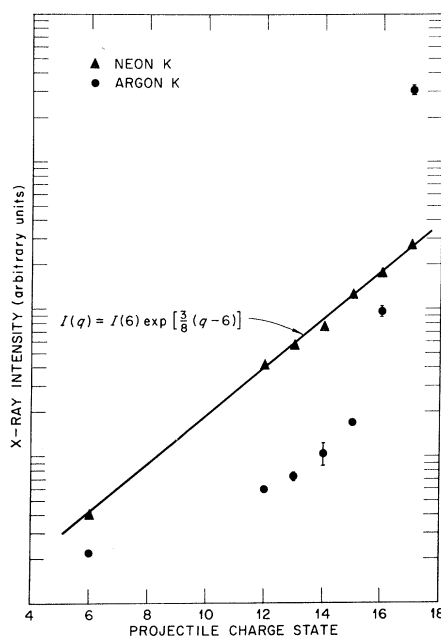


FIG. 2. Neon and argon K x-ray yield versus projectile charge state. The data have been normalized to the same gas-cell pressure and to the same number of incident projectiles. In all cases the statistical counting errors are smaller than the point symbol, as is the scatter of data from run to run except where otherwise indicated.

the decay of highly excited states of one- and two-electron neon ions. The production of excited states of one- and two-electron argon is also evident when the projectile charge state is +16 or +17.

Figure 2 presents the basic results of this experiment. It shows that both target and projectile K x-ray production depend strongly on the charge state of the projectile. Contrary to simple theories of x-ray production by direct ionization, which lead to a quadratic projectile charge dependence, the neon yield is found to fit the empirical expression

$$I(q) = I(6) \exp\left[\frac{3}{8}(q-6)\right],$$

where $I(q)$ is the intensity of the neon K line when the projectile has charge $+q$. While we know of no physical justification for this simple exponential behavior, it does emphasize that the behavior of target and projectile yields is *qualitatively* different, since the smoothly varying argon K-yield growth is faster than exponential.

The present method provides a means of tuning the projectile energy levels through those of the

target. Low charge states like Ar^{6+} have L shells whose energies lie above neon K levels. These levels swap near charge state +14 and for Ar^{16+} and Ar^{17+} the mismatch between the neon K and argon L levels is greater than 100 eV. If a level-matching mechanism of the type discussed above and in Ref. 1 were operative, we would expect that the neon yield would peak near projectile charge +14 and diminish at higher and lower charge states.⁴ The data presented in Fig. 2 show no evidence for a local enhancement near charge state +14, and we conclude that either sufficiently close level matching does not occur or that level matching is not the dominant mechanism for the projectile charge-state-dependent effect at this beam velocity.

An alternative explanation for this effect is based on the possibility that the electronic configuration of the target ion following a collision is strongly dependent upon the projectile charge state, and that more highly charged projectiles produce significantly more target ionization.⁵ More highly charged neon configurations should have a larger probability of emitting an x ray than less highly charged configurations.^{6,7} The fluorescence yield of neon varies from 0.018 for neon possessing a single vacancy⁸ in the K shell to 1.0 for the one-electron ion excited to the $2^2P_{1/2}$ state; the variation is most rapid for the highest charge states. From the crude assumption that K -shell vacancy production in neon is completely independent of projectile structure and that L -shell ionization is strongly dependent upon projectile structure, it follows that variations in fluorescence yield alone could account for a 56-fold increase in x-ray yield. Figure 2 shows that the yield does increase by a factor of 43 between projectile charge state +6 and +16 with a further additional increase at +17. One difficulty with this interpretation is that the more highly stripped the neon becomes, the higher should be the K x-ray energy.^{7,9} Our data show no evidence for a centroid shift until the projectile charge state reaches +17.

In the frame of the projectile, the collision is viewed as a neon atom incident on an ionized argon target. The binary-encounter model¹⁰ predicts that, for a bare neon projectile, the Ar K -shell ionization cross section should *decrease* with increasing Ar charge state due to the increased K -shell binding energy. The eightfold *increase* in argon K x-ray production between charge states +6 and +15 might thus also be attributed to a variation in fluorescence yield,¹¹

since an increase from ~ 0.122 to 1.0 is conceivable in going from a configuration with L shell full to one with L shell empty. The steep rise in production at charge state +15 and beyond can possibly be attributed to electron capture events in which captured electrons cascade through the $2p$ level into K -shell vacancies that already exist in metastable configurations of two- and three-electron ions and in all configurations of the one-electron ion. The large rate is attributed to the fact that electron capture cross sections in similar collision processes are 1 to 2 orders of magnitude larger than K -shell ionization cross sections.²

The probable creation during the collision of multiply ionized neon of unknown charge-state distribution and radiating configuration makes it difficult to determine an unambiguous inner-shell ionization cross section, even though the energies^{7,9} and fluorescence yields⁷ for many configurations have been estimated. It is possible, however, to set a lower limit on the magnitude of the neon K -shell ionization cross section σ_{ion}^K . The effective fluorescence yield $\bar{\omega}_K$ will be in all cases ≤ 1 . From our yield obtained with projectile Ar^{17+} we find $\bar{\omega}_K \sigma_{\text{ion}}^K(+17) \sim 8 \times 10^{-18} \text{ cm}^2$. Hence, for that projectile, $\sigma_{\text{ion}}^K(+17) \geq 8 \times 10^{-18} \text{ cm}^2$. This cross section is $\sim \frac{1}{5}$ as large as the cross section $4 \times 10^{-17} \text{ cm}^2$ computed by a binary-encounter model¹⁰ for a completely stripped argon projectile.

The present results indicate that experiments with sufficient resolution to resolve the individual neon and argon lines belonging to specific charge states are clearly needed to elucidate the large effects reported here. The evidence for the occurrence of excited neon lines near the one-electron Lyman-series limit was to us a considerable surprise and certainly merits further investigation.

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¹J. R. Mowat, D. J. Pegg, R. S. Peterson, P. M. Griffin, and I. A. Sellin, Phys. Rev. Lett. **29**, 1577 (1972).

²J. R. Macdonald, L. Winters, M. D. Brown, T. Chiao, and L. D. Ellsworth, Phys. Rev. Lett. **29**, 1291 (1972).

³W. Brandt, R. Laubert, M. Mourino, and A. Schwarzschild, Phys. Rev. Lett. **30**, 358 (1973).

⁴T. M. Kavanagh, M. E. Cunningham, R. C. Der, R. J.

Fortner, J. M. Khan, E. J. Zaharias, and J. D. Garcia, *Phys. Rev. Lett.* **25**, 1473 (1970).

⁵C. F. Moore, M. Senglaub, B. Johnson, and P. Richard, *Phys. Lett.* **40A**, 107 (1972).

⁶D. Burch, W. B. Ingalls, J. S. Rislely, and R. Heffner, *Phys. Rev. Lett.* **29**, 1719 (1972).

⁷C. P. Bhalla and M. Hein, *Phys. Rev. Lett.* **30**, 39 (1973).

⁸W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, *Rev. Mod. Phys.* **44**, 716 (1972).

⁹F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, 14 (1971).

¹⁰J. D. Garcia, *Phys. Rev. A* **4**, 955 (1971).

¹¹F. P. Larkins, *J. Phys. B: Proc. Phys. Soc., London* **4**, L29 (1971).

Nuclear Quadrupole Interactions at Te¹²⁵ in the Isoelectronic Crystalline Hosts of S, Se, and Te†

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A systematic increase in Te¹²⁵ quadrupole splitting of up to 54% has been observed in the isoelectronic crystalline hosts of Te, Se, and S. This is attributed to nonlocal and overlap electronic contributions to the electric field gradient q_{el} , which increase as a result of a systematic shortening of the covalent bonds.

We have observed large and unique quadrupole splittings (QS) at Te¹²⁵ in the isoelectronic hosts of orthorhombic S and trigonal Se using the 35.5-keV γ resonance.¹ In conjunction with the measured QS in Te metal,^{2,3} these data show a systematic increase of up to 54% in the QS which is found to correlate linearly with the inverse cube of the covalent bond lengths⁴ R that Te forms with its two near neighbors in these hosts.

These results are significant, as they will permit an understanding of the origin of the electric field gradient (EFG) in Te metal. It has been pointed out⁵ that for semimetals where the density of electron states at the Fermi level is small, the principal mechanism for the origin of the EFG is electronic and comes from all the band electrons below the Fermi surface. For the case of Sb metal where detailed calculations⁶ have been made, q_{el} is composed of local, nonlocal, and overlap contributions. Te and Sb both have partially filled 5*p* atomic shells and, in the solid state, are characterized as semimetals. The present results on Te¹²⁵ permit estimating the relative magnitudes of the different contributions to q . In particular these data reveal the existence of sizable contributions of q_{el} (overlap) and q_{el} (nonlocal) and further also suggest that q_{ion} and q_{el} (local) have opposite signs in Te metal. The study of EFG in metals remains a subject of continued interest both experimentally^{7,8} and theoretically.⁹ The present approach of using isoelectronic hosts in the study of quadrupole

interactions using nuclear γ resonance is novel and in a sense complements measurements made using other techniques^{10,11} as a function of temperature and pressure. The present measurements are significant in another way. They provide a calibration of covalent bond lengths in terms of QS in Te-based twofold coordinated hosts. This result is of interest in the structure determination of some amorphous semiconductors such as GeTe¹² and Se.¹³

2.7-yr Sb¹²⁵ in Cu was used as a source of monoenergetic γ rays of energy 35.5 keV. The source was prepared by diffusing carrier-free Sb¹²⁵ in high-purity Cu foils. A velocity spectrum of the source taken with a thin ZnTe absorber [Fig. 1(a)] gave an observed full width at half-maximum (FWHM) of 5.95 ± 0.08 mm/sec. In a forthcoming publication¹⁴ we will present details of linewidth measurements using ZnTe absorbers of different thicknesses. The linewidth of the Sb¹²⁵(Cu) source and the "f" of ZnTe at 78 K were found to be 5.10 ± 0.08 mm/sec and 0.32 ± 0.02 , respectively. The linewidth of these sources agrees with $2\Gamma_n = \hbar/\bar{\tau} = 5.20 \pm 0.04$ mm/sec based on the electronically measured lifetime¹⁵ of the 35.5-keV state. Alloys of the compositions Te_{0.02}Se_{0.98} and Te_{0.014}S_{0.986} were prepared by melting the elements *in vacuo*, followed by annealing the melt. The Se alloys were annealed at 150°C for 24 h and the S alloys at 50°C for about 6 h. X-ray analysis of the alloys confirmed the trigonal and orthorhombic phases of